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APPLICATION OF:) POLYESTER RESIN FOAMED ARTICLE
Takashi FUJIMAKI) AND PRODUCING METHOD THEREFOR
Yoshitomo URATA)

SERIAL No. 09/787,627) GROUP ART UNIT: 1732
FILED: Oct. 2, 1998) EXAMINER: Allan R. Kuhns

HONORABLE COMMISSIONER OF
PATENTS AND TRADEMARKS
WASHINGTON, D.C. 20231

SIR:

DECLARATION UNDER RULE 132

TAKASHI FUJIMAKI, being one of the inventors of the above identified application and a citizen of Japan residing at 634-4-442, Noba-cho, Kohnan-ku, Yokohama, 234-0056, Japan, does hereby declare the following facts pertinent to the application namely, that:

1. He graduated from Kyoto University, Faculty of Engineering, and completed Master of course specializing in Synthesis of Organometallic compounds & their catalytic functions.
2. From 1961 to 1997 he worked for Showa Denko K.K. and Showa Highpolymer Co., Ltd, a subsidiary of Showa Denko K.K. where he worked as a chief researcher in the R & D center, especially as an expert in polyolefins and biodegradable polyesters.
3. He is familiar with the related arts of saturated polyester and the references cited by the Examiner in

connection with the captioned application.

4. Experiments were conducted under his supervision in order to provide a comparison data regarding the effects of coupling agents for optimizing the MFR & swell of recycled polyethylene-terephthalate (PET).

5.1 Purport of the experiments

To confirm the synergistic effects of employing di-functional epoxy compound (D) and tri-functional epoxy compound (T) together as coupling agents.

5.2 Materials used in the experiments:

(a) Recycled PET bottles

Clear flakes commercially available from WITH PET BOTTLE CO., LTD:

Inherent viscosity: 0.73dl/g; Weight average molecular weight: 27,800;

Molecular weight distribution (Mw/Mn): 2.2;

MFR*: 60g/10min, Swell*: -21% (* after hot air drying for 12hr at 120.)

(b) Coupling agent

di-functional epoxide compound (D): ethylene glycol diglycidyl ether available

from Kyoeisha-Kagaku Co., Ltd. as EPOLITE 40E

tri-functional epoxide compound (T): trimethylol propane triglycidyl ether

available from Kyoeisha-Kagaku Co., Ltd. as EPOLITE 100MF

(c) Coupling reaction catalyst

Powdery mixture containing 50wt% of Ca-stearate, 25 wt% of Na-stearate,

12.5wt% of Li-stearate, and 12.5wt% of Mn-acetate.

(d) Twin-screw extruder

Ikegai Corp.,: PCM-70 (70mm ., L/D=37, 3 vent type)

5.3 Method for testing:

To 100 weight parts of non-dried PET flakes (a), 0.1 weight parts of coupling reaction catalyst and coupling agent (c) consisting of EPOLITE 40E and EPOLITE 100MF ratios of which are shown in Table 1 are added, and mixed in a Henschel mixer for 5 minutes.

This mixture of PET flakes was kneaded and reacted in a twin-screw extruder at 100 rpm and 280°C, vacuum degree of from -0.090 to -0.098 Mpa and 5 strands were extruded through nozzles of 2mm., quenched with water and cut by rotary cutter to make pellets. The resulting pellets were dried at 120°C, for 12 hours by hot air, then stored in a humidity proof bag.

MFR (280°C, load 2.16kg) and swell (rate of expansion) of the produced pellets were measured. Results are listed in Table 1 and plotted in Fig. 1, Fig. 2 and Fig. 3.

5.4 Evaluation and Considerations:

High molecular weight PET treated according to the present invention has a significantly reduced MFR (Fig. 1) and significantly increased swell (Fig. 2). These results are decisively affected by T/D ratio of the coupling.

Figure 3 shows a relationship between MFR and swell in terms of T/D ratio.

As can be seen from Fig. 3, the higher the T/D ratio, the greater the swell and the lower the MFR. As the result, pellets having a MFR of 10 or lower which is suitable for making a tubular film or direct molding of blown bottles can easily be obtained.

When only ethyleneglycol diglycidylether which is a difunctional epoxy compound is used, melt viscosity does not increase and MFR barely becomes 10 or lower, as it has a linear structure similar to conventional commercial PET. Therefore, it cannot be appropriately employed for making tubular film and direct molding for blown bottles.

On the other hand, when used with trimethylolpropane triglycidylether which is a tri-functional epoxy compound, MFR and swell are easily controlled by introducing long chain branched structures through adjustment of the T/D ratio. Thus, new molded articles which cannot be made by linear PET as commercially obtained can be developed.

He hereby declares that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further he is hereby warned that willful, false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of title 18 of the United States Code and that such willful, false statements may jeopardize the validity of the application or any registration resulting therefrom.

Takashi Fujimaki

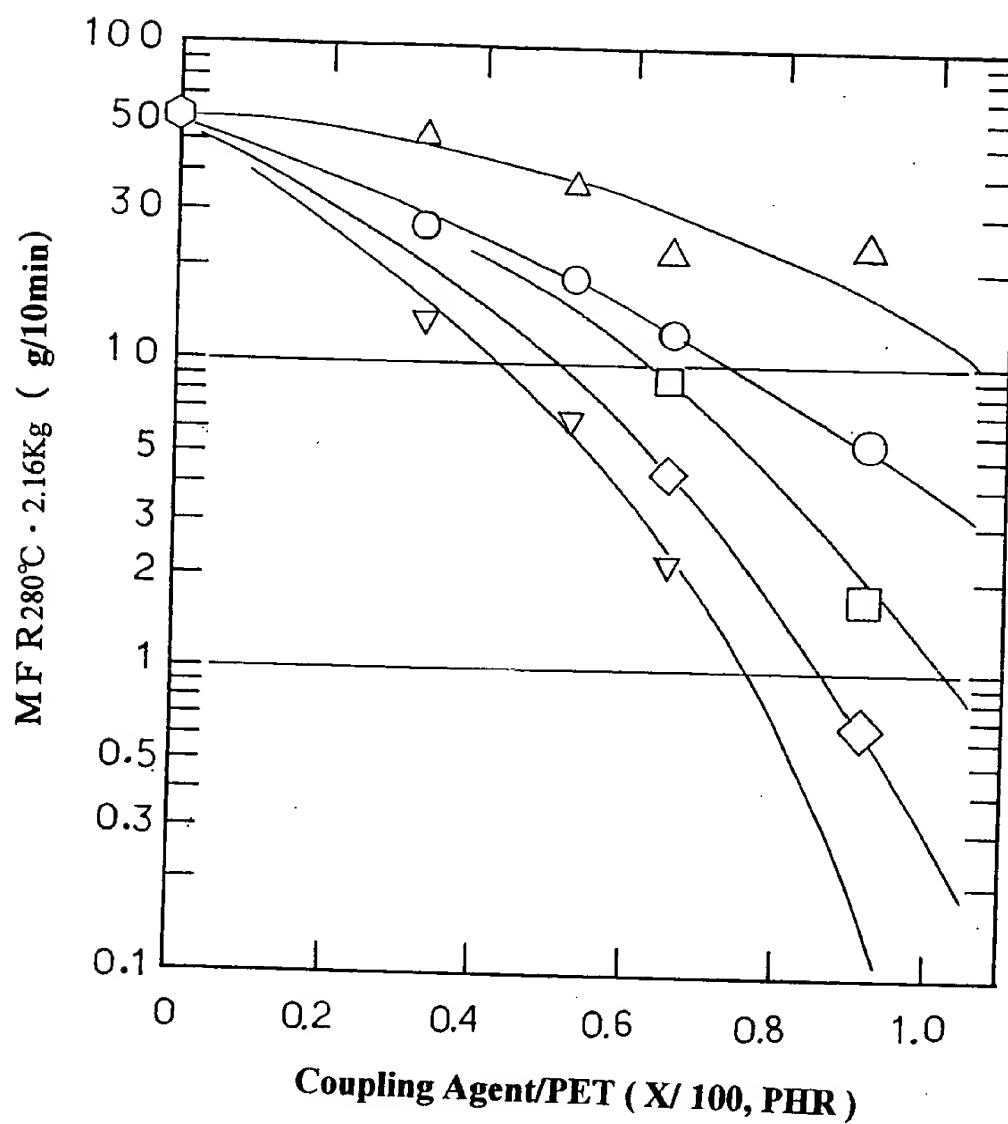
TAKASHI FUJIMAKI

Date: January 9, 2004.

Table 1

No.	Composition PET/Coupling agent/Cat	T/D ratio (wt. part)	MFR (g/10min)	Swell (%)	Symbol in Figures
1-1	100/ 0.33/ 0.1	0/100	50	-5	△
1-2	100/ 0.52/ 0.1	0/100	37	3	△
1-3	100/ 0.65/ 0.1	0/100	22	12	△
1-4	100/ 0.91/ 0.1	0/100	24	8	△
2-1	100/ 0.33/ 0.1	25/ 75	27	26	○
2-2	100/ 0.52/ 0.1	25/ 75	19	32	○
2-3	100/ 0.65/ 0.1	25/ 75	12	55	○
2-4	100/ 0.91/ 0.1	25/ 75	5.7	72	○
3-1	100/ 0.65/ 0.1	50/ 50	9.1	75	□
3-2	100/ 0.91/ 0.1	50/ 50	1.7	120	□
4-1	100/ 0.65/ 0.1	75/ 25	4.5	110	◇
4-2	100/ 0.91/ 0.1	75/ 25	0.67	190	◇
5-1	100/ 0.33/ 0.1	100/ 0	13	90	▽
5-2	100/ 0.52/ 0.1	100/ 0	6.5	125	▽
5-3	100/ 0.65/ 0.1	100/ 0	2.2	160	▽

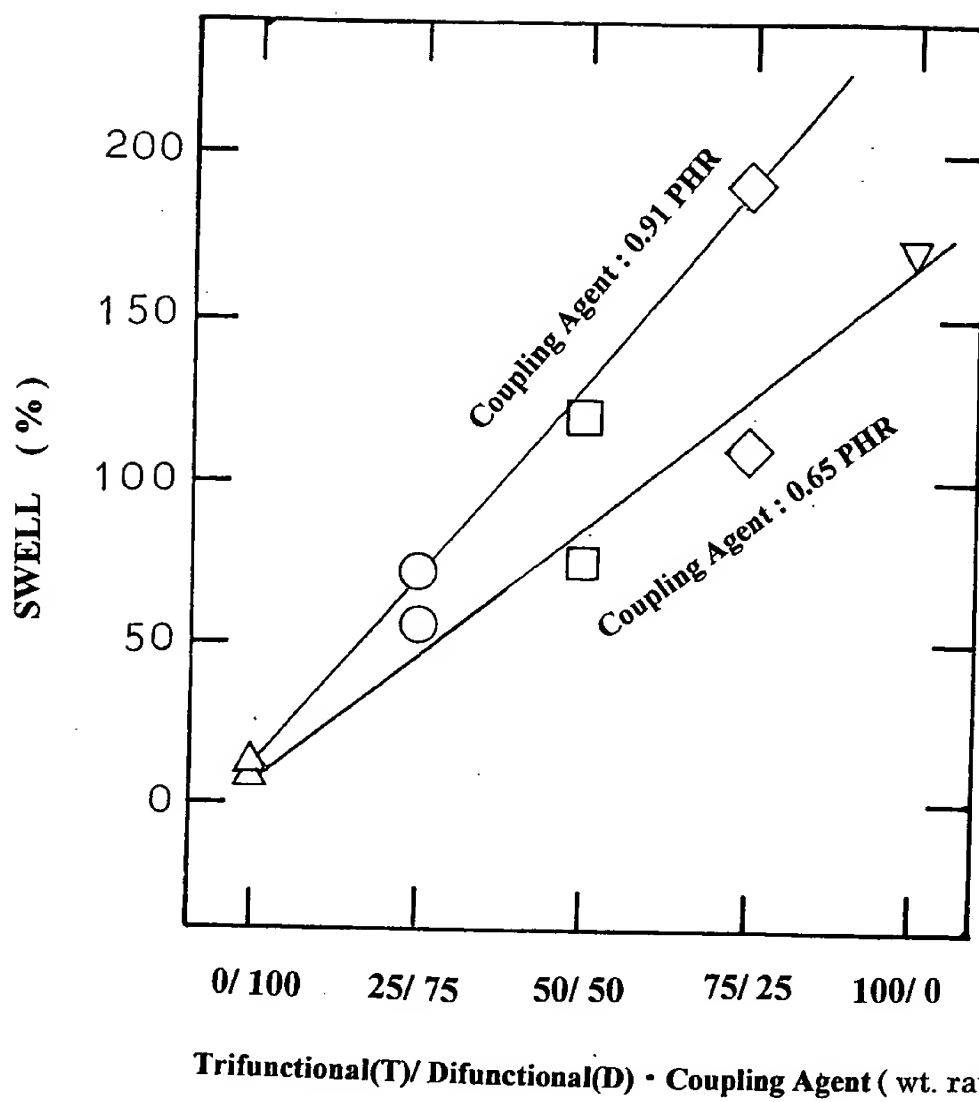
Fig. 1



(Symbol for T/D ratio)

0/ 100	△
25/ 75	○
50/ 50	□
75/ 25	◇
100/ 0	▽

Fig. 2



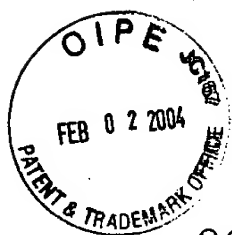


Fig. 3

